Preparation and Characterization of Small Sized PZT Powders: A Sol-Gel Modified Approach

Alka, Yong woo choi, Oh H. Kwon and Kyu M. Choi*
Department of Electronics & Information Communication Engineering
Kwandong University, Yangyang, Kangwondo 215-802 (Korea)

Abstract

A propyl alcohol based sol-gel method was used as to replace the 2-methoxyethanol with 1,1,1 tris (hydroxymethyl) ethane for preparation of pzt piezoelectric ceramic.. The powder obtained by this sol-gel process were calcined at 900 ^{0}C and followed by the sintering at 1100 ^{0}C for *ca*2 hrs as to reach a pyrochlore-free crystal phase. The characterization of synthesized material was carried out by the XRD analysis and the surface morphology were determined by high resolution scanning electronmicroscopy. Further, the prepared small sized pzt thin powders are likely to be used in various applications.

Key words: PZT, sol-gel

Introduction

Due to their distinctive energy conversion function, lead zirconate titnate (PZT) based ceramics are used for sensors, transducers, and capacitors[1,2] Several methods were presented for the preparation of the PZT powders viz., hydrothermal solutions, solid state reaction, spin-coating and sol-gel etc. [3].A propyl alcohal based sol-gel method was used to prepare the nanoscale PZT powders[4]. The present investigation deals the detailed method of the preparation of the small sized PZT powders as applying by various firing condition and then to determine its resistivity towards the temperature.

Experimental Procedure

PZT precursor solution of the composition $Pb_1Zr_{.53}Ti_{.47}$ was prepared from the $Pb(CH_3CO_2)_2.3H_2O$(lead acetate trihydrate), $Zr(C_3H_7O)_4$(zirconium tetra propoxide). 70% wt solution in1-proponal and $Ti[(CH_3)_2CHO]_4$(Titanium (IV)isopropoxide) 97%[8] Lead acetate was dissolved into the propyl alchol by the ratio 15:1. Dehydare the solution at 110 ^{0}C and then reflux it under stirring for 2 hrs at the same temperature. Cooled down the solution mixture to 90 ^{0}C and then to add $Zr(C_3H_7O)_4$ and glacial acetic acid to check the pH as to be kept constant as about ~5. Again stirred the solution and reflux at 110^{0}C for ca 1h. Further, in this lead zircounium solution, added $Ti[(CH_3)_2CHO]_4$ solution Stirred and reflux the solution mixture for ca 2 hrs at 110^{0}C a clear yellow PZT solution was obtained which was returned to gel form. while cooled down at room temperature.

*Corresponding author E.Mail: kmchoi@kd.ac.kr

Further, the gel was evaporated at 120 ^{0}C to obtained yellowish white PZT powders. The powder was calcined at 900 ^{0}C for various time intervals i.e., from 1 hrs to 4 hrs in the alumina crucible. The calcined powders were further subjected for the preparation of disk. These disks were encapsulated in alumina crucible to avoid any loss of lead in sintering process as was then sintered at 1000 ^{0}c, 1050 ^{0}c, 1100 ^{0}C, 1150 ^{0}C, 1200 ^{0}C. Now we can obtain the PZT Ceramic. The crystal phase was determined by the X-ray diffraction method by using the Rigakeu (D-max-2200) X-ray diffraction meter (XRD). Moreover, the surface morphology was analyzed by using high-resolution scanning electron microscope (SEM). Ag polished the PZT ceramics cleaned and coating.. The modified Sawyer Tower circuit [9] was used to ascertain the ferroelectric property of PZT ceramic. HEWLETT PACKARD 4284A was used for determining the dielectric constant and dielectric losses.

Result and discussion

The X-ray diffraction analysis obtained for calcined powders at 900^{0}C for 1h, 2h, 3h, 4h are shown in fig1. The fig clearly demonstrate that the amount of pyrochlore phase was gradually decreased with increasing the calcining period as after 4h of calcining no pyrochlore phase was existed, only the perovskite phase occurred. when the calcining time was 2h or more, the (100), (200) and (211) planes were splited into two peaks. Hence, it can be concluded that the tetragonal phase and rhombohedral phase coexisted during longer calcining period. The SEM images for these PZT ceramics sintered at various temperatures were returned in figure 2

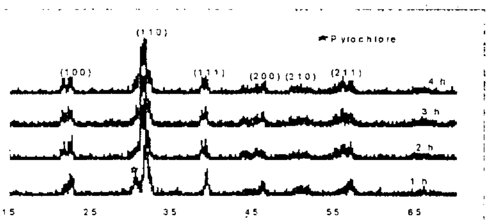
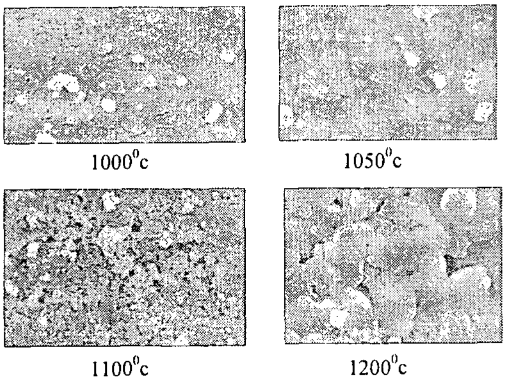


Fig (1) X-ray diffraction patterns for PZT powders calcined at 900^{0}C as a function of calcining time.

It was to be noted that reasonably a good homogeneous grain distribution was obtained for the PZT ceramics when sintered at 1100 ^{0}C and posses minimum average grain size.



Fig(2) 900^{0}C for 4h and sintered at 1000^{0}C, 1050^{0}C SEM of PZT ceramic calcined at, 1100^{0}C, 1200^{0}C for 2h.

and minimum dielectric loss. The device size and the quality factor are inverse proportional to the ${}_r$ and the However, at lower sintering temperature i.e., 1000 ^{0}C average grain sizes are big as may be due to the reaction was incomplete. Moreover, at higher sintering temperature the grain size was larger Further, the grain size was estimated at 1100^{0}C and it was found to in the range of 400 to 500 nm . Moreover, when it was sintered at 1100 ^{0}C, 2h showed maximum dielectric constant i.e., 880 and showed minimum dielectric loss. The device size and the quality factor are inverse proportional to the ${}_r$ and the dielectric loss respectively [6].. The above studies enabled that the PZT powder calcined at 900^{0}C for 4h, and sintered at 1100 ^{0}C for 2h were chosen for further assessment of

electrical properties.The ferroelectricity showed the hysteresis loop which was obtained by the modified shower tower circuit. Poling voltage used were 3 kV mm^{-1}, applied for 20 min at 120 ^{0}C [10]. The polarization and electric field are 4.27C/cm^2 and 15.6 kV/cm per division respectively

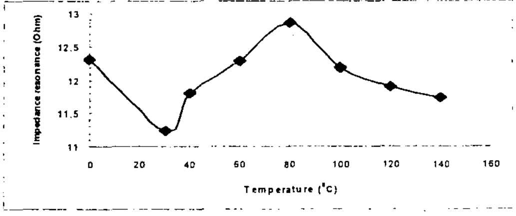


Figure 3:Impedance as a function of temperature

Fig3 shows that at the beginning at room temperature resistance is low and produce a large current after that increasing the temperature the resistance increases drastically hence the current will be reduce. Therefore, a smart self-regulating heating circuit is found. Smart structures are an integration of sensors, actuators, and control system [5].

Conclusions

The sol-gel method is mainly affected by the firing conditions and must use the excess of Pb to recover the loss of Pb during the sintering process. XRD data showed that the pyrochlore phase was completely disappeared at the calcining time 4h. Dielectric constant dielectric loss, resonance frequency,and temperature dependence Impedance were measured. We found that the PZT samples have good piezoelectric property and will have the potential for application such as various piezoelectric transducers and sensors.

Acknowledgement

This work was supported by grant No. RTI05-01-02 from the Regional Technology Innovation Program of the Ministry of Commerce, Industry and Energy(MOCIE)

Reference

1. K. Uchino,Am. Ceram.Soc. Bull.65(1986) 647-652
2. Q. M Wang, L.E. Cross, J.Am. Ceram. Soc.82(1990) 1641-1643
3. T. Tanase, Y. Kobayashi, M. Konno Thin solid film 457(2004)264-267.
4. C. L Huang, B. H. Chen, L. Wu. Materials Reserch Bull. 39(2004)523-532
5. Proc. Natl. Acad. Sci. USA Vol. 96, pp 8330-8331, July1999.
6. . H. Zhao.,T Ren, N. Zhang, ,material science and engineering B99 (2003)192-194.